

# CONTENTS

PREFACE	v
ORGANIZATION OF THIS BOOK	ix
CONTRIBUTORS	xi
<b>CHAPTER 1</b>	
<b>DIMENSIONALITY AND OPTICAL RESPONSES OF MATERIALS</b>	<b>1</b>
<i>T. Ogawa</i>	
1.1. INTRODUCTION	2
1.2. DIMENSIONALITY IN THE SINGLE-ELECTRON STATES	3
1.2.1. <i>Density of States of Bloch Electrons</i>	4
1.2.2. <i>Band Structures of a Quasi-One-Dimensional System: Comparison between a Bulk Crystal and a Wire-Shaped Crystal of Silicon</i>	4
1.2.3. <i>Topological Features of Artificial Low-Dimensional Structures</i>	7
1.3. DIMENSIONALITY IN THE WANNIER EXCITON STATES	9
1.3.1. <i>Excitonic Effects on One-Photon Absorption Processes in <math>d</math> Dimension</i>	9
1.3.1.1. <i>The three-dimensional case</i>	11
1.3.1.2. <i>The two-dimensional case</i>	12
1.3.1.3. <i>The one-dimensional case</i>	15
1.3.1.4. <i>The zero-dimensional case</i>	21
1.3.2. <i>Excitonic Effects on Two-Photon Absorption Processes in <math>d</math> Dimension</i>	23
1.3.2.1. <i>The three-dimensional case</i>	23
1.3.2.2. <i>The two-dimensional case</i>	24
1.3.2.3. <i>The one-dimensional case</i>	25
1.3.2.4. <i>The zero-dimensional case</i>	27
1.3.2.5. <i>Origins of the TPA anisotropy</i>	27
1.3.3. <i>A Two-Photon Absorption Experiment in GaAs Quantum Wires</i>	28
1.3.4. <i>Dimensional Crossover of Excitons</i>	30
1.3.4.1. <i>A Q2D exciton with a quantized center-of-mass motion</i>	31
1.3.4.2. <i>Exciton states in an intermediate dimension</i>	32
1.3.4.3. <i>TPA spectra in an intermediate dimension</i>	33
1.3.5. <i>Geometrical Effects on the Exciton Center-of-Mass Motion</i>	34
1.3.6. <i>Exciton Confinement on a Spherical Shell</i>	35
1.3.7. <i>Excitonic Molecules in One Dimension</i>	36
1.3.7.1. <i>Heitler-London scheme for 1D excitonic molecule</i>	36
1.3.7.2. <i>Four-fermion problem for an excitonic molecule on a 1D lattice</i>	37
1.4. DIMENSIONALITY IN THE DIELECTRIC CONFINEMENT EFFECTS	39
1.4.1. <i>Quasi-Two-Dimensional Systems</i>	39

1.4.2. Quasi-One-Dimensional Systems	41
1.5. DIMENSIONALITY IN THE FERMI-SURFACE EFFECTS	43
1.5.1. The Peierls Instability	43
1.5.2. The Tomonaga-Luttinger Liquid	45
1.5.3. Fermi-Edge Singularity in Optical Processes	49
1.5.3.1. FES in the single-band Tomonaga-Luttinger liquids	50
1.5.3.2. An experimental observation of the FES in Q1D	53
1.5.3.3. FES in the spin-split Tomonaga-Luttinger liquids	54
1.5.3.4. FES in the two-band Tomonaga-Luttinger liquids	58
1.6. DIMENSIONALITY IN THE SELF TRAPPING	65
1.6.1. Overview of the Dimensionality in the Self Trapping	65
1.6.2. A Unified Treatment of the Self Trapping in $d$ Dimension	67
1.6.2.1. Self trapping in one dimension	68
1.6.2.2. Self trapping in two dimension	69
1.6.2.3. Self trapping in three dimension	70
1.7. DIMENSIONALITY IN THE PHOTOINDUCED STRUCTURE CHANGES	72
1.7.1. Nucleation Picture	73
1.7.2. Mean-Field Picture	76
1.8. CONCLUDING REMARKS	80
REFERENCES	83

## CHAPTER 2

### ***Ab initio* CALCULATION OF NONLINEAR OPTICAL SUSCEPTIBILITY**

*T. Nakayama*

2.1. INTRODUCTION	91
2.1.1. Purpose of This Chapter	91
2.1.2. Microscopic Nonlinear Optical Susceptibility	94
2.1.3. Nonlinear Optical Phenomena	99
2.1.3.1. Conservative nature in $\chi^{(n)}$	99
2.1.3.2. Complex nature in $\chi^{(n)}$	101
2.2. METHOD OF <i>ab initio</i> CALCULATION	104
2.2.1. <i>Ab initio</i> Band Structure Calculations	104
2.2.2. Band-State Expression of $\tilde{\chi}^{(n)}$	106
2.2.3. Momentum Matrix Element	110
2.2.3.1. Momentum renormalization	110
2.2.3.2. Numerical aspects	111
2.2.4. Numerical Convergence in $\chi^{(n)}$ Calculation	115
2.3. TWO-PHOTON ABSORPTION IN BULK SEMICONDUCTORS	117
2.3.1. Two-Photon-Absorption Coefficient	117
2.3.2. Degenerate TPA Spectra	119
2.3.3. Non-Degenerate TPA Spectra	124
2.3.3.1. TPA anisotropy	126

2.4. VARIOUS NONLINEAR SPECTRA IN BULKS AND SUPERLATTICES	129
2.4.1. <i>Second and Third Harmonic Generations in Bulk Semiconductors</i>	129
2.4.1.1. <i>SHG spectra</i>	129
2.4.1.2. <i>THG spectra</i>	131
2.4.2. <i>TPA and SHG Spectra in Semiconductor Superlattices</i>	132
2.4.2.1. <i>Anisotropy in TPA spectra</i>	134
2.4.2.2. <i>Anisotropy in SHG spectra</i>	134
2.5. FUTURE PROSPECTS	137
2.5.1. <i>A Future Prospect</i>	137
2.5.2. <i>Local Field Effects</i>	138
2.5.3. <i>Excitonic Effects</i>	141
REFERENCES	143

### CHAPTER 3

## WANNIER-STARK LOCALIZATION IN SEMICONDUCTOR SUPERLATTICES

147

*M. Nakayama*

3.1. INTRODUCTION	148
3.2. MINIBAND STRUCTURES	150
3.2.1. <i>Band Discontinuity</i>	150
3.2.2. <i>Envelope-Function Approximation</i>	152
3.3. THEORETICAL MODELS OF WANNIER-STARK LOCALIZATION	156
3.3.1. <i>Nearest-Neighbor Tight-Binding Model</i>	156
3.3.2. <i>Transfer-Matrix Method</i>	159
3.4. STARK-LADDER STATES	161
3.4.1. <i>Sample Structures and Spectroscopic Methods</i>	161
3.4.2. <i>Transformation Process from Miniband States to Stark-Ladder States</i>	165
3.4.3. <i>Exciton Binding Energy</i>	172
3.4.4. <i>Oscillator Strengths of Stark-Ladder Transitions</i>	173
3.4.5. <i>Stark-Ladder Transitions in Type-II Superlattices</i>	176
3.4.6. <i>Electro-Optical Bistability</i>	176
3.5. RESONANT COUPLING BETWEEN STARK-LADDER STATES	177
3.5.1. <i>Anticrossing between Stark-Ladder Levels</i>	177
3.5.2. <i>Wave-Function Delocalization</i>	183
3.6. ABOVE-BARRIER STATES	185
3.6.1. <i>Existence of Above-Barrier States</i>	185
3.6.2. <i>Stark-Ladder Formation and Resonant Coupling</i>	188
3.7. BLOCH OSCILLATIONS	192
3.7.1. <i>Concepts of Bloch Oscillations</i>	192
3.7.2. <i>Observation of Bloch Oscillations</i>	193
3.8. SUMMARY	195
REFERENCES	196

<b>CHAPTER 4</b>	
<b>ULTRAVIOLET LASER EMISSION FROM ZnS-BASED QUANTUM WELLS</b>	202
<i>Y. Yamada</i>	
4.1. INTRODUCTION	202
4.2. MOCVD GROWTH AND STRUCTURES	204
4.3. BAND LINEUPS IN STRAINED-LAYER SYSTEMS	205
4.3.1. <i>Effects of Strains on Interface Structures</i>	207
4.3.2. <i>Derivation of Band Lineups</i>	208
4.3.3. <i>Band Offsets in Cd<sub>x</sub>Zn<sub>1-x</sub>S-ZnS Systems</i>	209
4.4. FORMATION OF OPTICAL GAIN IN II-VI QUANTUM WELLS	214
4.4.1. <i>Experimental Procedures</i>	214
4.4.2. <i>Ultraviolet Stimulated Emission under Optical Pumping</i>	216
4.4.3. <i>Ultraviolet Injection Laser Diode</i>	221
4.4.4. <i>Optical Gain Spectra</i>	222
4.4.5. <i>Discussion</i>	225
4.5. CONCLUSIONS	235
REFERENCES	236
<b>CHAPTER 5</b>	
<b>LUMINESCENCE FROM SILICON NANOSTRUCTURES</b>	240
<i>Y. Kanemitsu</i>	
5.1. INTRODUCTION	240
5.2. CLUSTERS AND POLYMERS: FROM MOLECULES TO NANOSTRUCTURES	242
5.2.1. <i>Small Clusters</i>	243
5.2.2. <i>Polymers</i>	247
5.3. NANOCRYSTALLITES: FROM BULK TO NANOSTRUCTURES	253
5.3.1. <i>Fabrication of Nanocrystallites</i>	254
5.3.2. <i>Red Photoluminescence</i>	256
5.3.3. <i>Blue and Green Luminescence</i>	266
5.4. APPLICATIONS	274
5.4.1. <i>Light-Emitting Diodes</i>	274
5.4.2. <i>Optical Logic Gates</i>	278
5.5. SUMMARY	282
REFERENCES	283
<b>CHAPTER 6</b>	
<b>OPTICAL PROPERTIES OF Pb-BASED INORGANIC-ORGANIC PEROVSKITES</b>	288
<i>T. Ishihara</i>	

6.1. INTRODUCTION	289
6.2. MATERIALS AND STRUCTURES	290
6.2.1. Perovskite Compounds	290
6.2.2. Inorganic-Organic Perovskites	290
6.2.3. $C_{10}$ -PbI <sub>4</sub> (2D)	294
6.2.4. PhE-PbI <sub>4</sub> (2D)	296
6.2.5. PhE-Pb <sub>2</sub> I <sub>7</sub> ("2.5D")	296
6.2.6. C <sub>1</sub> -PbI <sub>3</sub> (3D)	297
6.2.7. C <sub>5</sub> -PbI <sub>3</sub> (1D)	297
6.2.8. C <sub>1</sub> -PbI <sub>6</sub> ·H <sub>2</sub> O (0D)	297
6.3. SAMPLE PREPARATION	298
6.3.1. Synthesis, Purification, and Crystal Growth	298
6.3.1.1. $C_{10}$ -PbI <sub>4</sub> (2D)	298
6.3.1.2. Other 2D compounds	298
6.3.1.3. C <sub>1</sub> -PbI <sub>3</sub> (3D) and C <sub>1</sub> -PbI <sub>6</sub> ·H <sub>2</sub> O (0D)	299
6.3.1.4. Spin coating	299
6.3.2. Cleavage	300
6.4. FUNDAMENTAL OPTICAL PROPERTIES	300
6.4.1. $C_{10}$ -PbI <sub>4</sub> (2D)	300
6.4.1.1. Electronic levels in the visible and ultraviolet region	300
6.4.1.2. Luminescence properties of $C_{10}$ -PbI <sub>4</sub>	303
6.4.2. C <sub>n</sub> -PbI <sub>4</sub> (2D)	304
6.4.3. C <sub>6</sub> -PbI <sub>4</sub> (2D)	307
6.4.4. PhE-PbI <sub>4</sub> (2D)	308
6.4.4.1. Electronic structure of PhE-PbI <sub>4</sub>	308
6.4.4.2. Photoconductivity	310
6.4.5. PhE-Pb <sub>2</sub> I <sub>7</sub> ("2.5D")	311
6.4.6. C <sub>1</sub> -PbI <sub>3</sub> (3D)	311
6.4.7. C <sub>1</sub> -PbI <sub>6</sub> ·H <sub>2</sub> O (0D)	312
6.4.8. C <sub>n</sub> -PbX <sub>4</sub> (X=Br,Cl) (2D)	313
6.4.9. C <sub>n</sub> -PbX <sub>4-x</sub> Y <sub>x</sub> (X,Y=Cl,Br,I) (2D)	314
6.4.10. C <sub>5</sub> -PbI <sub>3</sub> (1D)	314
6.4.11. Related Compounds	315
6.5. TOPICS IN Pb-BASED INORGANIC-ORGANIC PEROVSKITES	316
6.5.1. Models for Electronic Structure	316
6.5.1.1. Cationic model	316
6.5.1.2. Charge transfer model	316
6.5.2. Band Calculations	317
6.5.3. Unified Model for an nD Network of [PbI <sub>6</sub> ] <sup>4-</sup>	317
6.5.4. Image Charge Effect	321
6.5.4.1. Image charge effect on exciton binding energy	321
6.5.4.2. Image charge effect on the spectral position	325
6.5.5. Polariton Luminescence	326
6.5.5.1. Time-resolved luminescence of PhE-PbI <sub>4</sub>	327

6.5.5.2. <i>Temperature dependence of PL intensity</i>	328
6.5.6. <i>Pressure Effect</i>	330
6.5.7. <i>Raman Scattering and Phase Transition</i>	330
6.5.7.1. <i>Raman scattering</i>	330
6.5.7.2. <i>Structural phase transition and electronic states</i>	331
6.5.8. <i>Nonlinear Optical Properties</i>	332
6.5.8.1. <i>Third harmonic generation</i>	332
6.5.8.2. <i>High density optical excitation</i>	333
6.5.8.3. <i>Two-photon absorption</i>	334
6.5.9. <i>Transport and Electroluminescence</i>	334
6.6. CONCLUDING REMARKS	335
REFERENCES	336

## CHAPTER 7

### SOLID STATE PROPERTIES OF C<sub>60</sub> AND ITS RELATED MATERIALS

<i>Y. Iwasa</i>	340
7.1. INTRODUCTION	340
7.2. OPTICAL PROPERTIES OF UNDOPED FULLERENES	343
7.3. SPECTROSCOPIC STUDY OF ALKALI C <sub>60</sub> FULLERIDES	353
7.3.1. <i>Introduction</i>	353
7.3.2. <i>Optical Spectra</i>	355
7.3.3. <i>Discussion on A<sub>4</sub>C<sub>60</sub></i>	358
7.3.4. <i>Discussion on A<sub>3</sub>C<sub>60</sub></i>	361
7.4. C <sub>60</sub> POLYMERS	365
7.4.1. <i>Introduction</i>	365
7.4.2. <i>Photoinduced Polymerization</i>	366
7.4.3. <i>Polymerization in Alkali C<sub>60</sub> Fullerides</i>	367
7.4.4. <i>Pressure-Induced Polymerization</i>	369
7.4.5. <i>Discussion</i>	378
7.5. SUMMARY	379
REFERENCES	380

## CHAPTER 8

### ARRAYED NANOCCLUSERS IN ZEOLITE CRYSTALS

<i>Y. Nozue</i>	387
8.1. SPECTROSCOPIC STUDY OF NEW MATERIALS	387
8.1.1. <i>The Aim of Spectroscopic Study</i>	387
8.1.2. <i>Spectroscopic Study of Low-Dimensional Materials</i>	389
8.2. ZEOLITE CRYSTALS AS THE CONTAINER OF CLUSTER	390
8.3. SEMICONDUCTOR CLUSTERS	393

8.3.1. <i>Quantum Electronic States</i>	393
8.3.2. <i>Optical Properties of PbI<sub>2</sub> Clusters</i>	396
8.4. ALKALI METAL CLUSTERS	400
8.4.1. <i>Quantum Electronic States</i>	400
8.4.2. <i>Electron-Phonon Interaction</i>	408
8.4.3. <i>Magnetic Properties</i>	410
REFERENCES	412